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Well-to-Wheels analysis of hydrogen production from bio-oil reforming for use in internal combustion engines

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ABSTRACT

The environmental profile of hydrogen depends greatly on the nature of the feedstock and the production process. In this Well-to-Wheels (WTW) study, the environmental impacts of hydrogen production from lignocellulosic biomass via pyrolysis and subsequent steam reforming of bio-oil were evaluated and compared to the conventional production of hydrogen from natural gas steam reforming. Hydrogen was assumed to be used as transportation fuel in an internal combustion engine vehicle. Two scenarios for the provision of lignocellulosic biomass were considered: wood waste and dedicated willow cultivation. The WTW analysis showed that the production of bio-hydrogen consumes less fossil energy in the total lifecycle, mainly due to the renewable nature of the fuel that results in zero energy consumption in the combustion step. The total (fossil and renewable) energy demand is however higher compared to fossil hydrogen, due to the higher process energy demands and methanol used to stabilize bio-oil. Improvements could occur if these are sourced from renewable energy sources. The overall benefit of using a CO₂ neutral renewable feedstock for the production of hydrogen is unquestionable. In terms of global warming, production of hydrogen from biomass through pyrolysis and reforming results in major GHG emissions, ranging from 40% to 50%, depending on the biomass source. The use of cultivated biomass aggravates the GHG emissions balance, mainly due to the N₂O emissions at the cultivation step.

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1. Introduction

Global warming and the associated efforts for decreasing CO₂ emissions have rendered hydrogen one of the most important energy carriers of this century. Hydrogen can potentially be “CO₂-free” when used as fuel in fuel cells or internal combustion engines, as it does not contain carbon in its structure and thus

only H₂O is formed from its reaction with oxygen. However, hydrogen does not occur free in nature and has to be extracted from hydrogen-containing sources. Its environmental profile depends on both on the actual resource used (fossil vs renewable) and the process employed and therefore, it is essential to assess the different hydrogen production methods and power trains using a life cycle analysis or well-to-wheels approach.

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Life cycle assessment (LCA) and/or Well-to-Wheels (WTW) studies address the environmental aspects and potential impacts throughout a product's life, from raw material acquisition through production, use and disposal. Thus, they constitute valuable tools for assessment and identification of the most effective alternative feedstocks and processes, in order to achieve maximum benefits in sustainability and reduction of airborne emissions.

Currently, almost 95% of hydrogen is produced from fossil fuels such as natural gas, naphtha, heavy oil, and coal via steam reforming and partial oxidation processes [1]. Biomass has been proposed as an alternative feedstock for hydrogen production not only because it is renewable but also because it is a CO₂ neutral energy source. Hydrogen can be produced from biomass primarily via two thermo-chemical processes: gasification [2,3] and fast pyrolysis followed by steam reforming of pyrolysis oil [4–12]. Pyrolysis oil, known as bio-oil, is a complex mixture of acids, alcohols, aldehydes, esters, ketones, sugars, phenols, guaiacols, syringols, furans, and multifunctional compounds [13]. Bio-oil can be separated into a water-monomer-rich phase that contains mostly carbohydrate-derived compounds and a hydrophobic oligomer-phase composed mainly of lignin-derived oligomers [14]. To date, most research has been focused on the reforming of the aqueous fraction of bio-oil [4,5,7–12]. However, recent efforts are directed towards reforming the whole bio-oil, including also the oligomeric lignin-derived material [6,15].

The attractiveness of hydrogen lies not only in the variety of feedstocks and processes to produce hydrogen, but also the diversity of methods to produce energy from hydrogen (internal combustion engines, gas turbines, fuel cells) with virtually zero harmful emissions and potentially high efficiency at the point of use. Although there are still serious challenges to overcome for the use of hydrogen as transportation fuel (low energy density, on board vehicle storage, infrastructure etc), the advantages offered are significant enough to warrant the exploration of its possibilities. The use of hydrogen in fuel cell vehicles represents one of the most attractive options, offering high efficiency and zero emissions. The efficiency of fuel cell vehicles in practical driving conditions can be up to 60%, higher than the 45% efficiency achieved when hydrogen is used in an internal combustion engine (H₂-ICE) [16]. Still, fuel cell vehicles are still in the early stages of development, while the H₂-ICE vehicles represent a more realistic midterm option. According to Davis et al. [16], H₂-ICE vehicles will become available in the near term at a lower cost than fuel cell vehicles.

Several LCA and Well-to-Wheel studies have addressed the environmental impact of hydrogen, produced from several sources such as from natural gas via steam reforming [17–24], from electrolysis with electricity produced from nuclear and renewable energy sources [18,20,25,26], from biomass via gasification [18,20,27,28], and used in different power trains, such as in conventional ICE engine vehicles, fuel cell cars, hybrid electric vehicles [18,20] etc. To our best knowledge, an LCA or WTW analysis on the production of hydrogen from the steam reforming of bio-oil produced via biomass fast pyrolysis for use in a H₂-ICE vehicle has yet to be conducted. Only few reports were found, addressing techno-economic parameters and operating variables of the bio-oil reforming process investigated with Aspen [9,29–33].

The primary aim of this Well-to-Wheels study is to evaluate and quantify the environmental impacts of H₂ production from lignocellulosic biomass via pyrolysis and subsequent steam reforming of bio-oil for use in H₂-ICE vehicle. The WTW results of bio-hydrogen are compared to the respective environmental impacts of the conventional production of hydrogen via natural gas steam reforming for the same end-use, considered here as the reference case, as it is currently the most widely used technology for hydrogen production. An LCA study, as defined by the 14000 ISO standards, consists of four steps [34]: (1) Goal and scope definition; (2) Life cycle inventory (LCI) stage; (3) Life cycle impact assessment (LCIA) phase and (4) Interpretation of the results. The present WTW work was performed according to the four steps defined by the ISO standards, which are presented in detail in the following paragraphs.

2. Goal and scope of study

2.1. Goal

The present study focuses on the lifecycle environmental performance of H₂ produced from renewable (biomass) compared to that produced from fossil (natural gas) resources for use in H₂-ICE vehicle powertrain. Results are intended to be used as supporting data for the development of such hydrogen production technologies from biomass.

The investigated scenarios are the following:

Scenario A: Fast pyrolysis of lignocellulosic biomass for the production of bio-oil and subsequent steam reforming of bio-oil to hydrogen. Two types of feedstocks are considered: A1. wood waste, generally comprising of forest residuals, mill residues (e.g. bark and other wastes of pulp mills), secondary wastes from the wood industry (sawdust, shavings etc) and forest litter and A2. dedicated willow plantations (fast-growing energy crop exhibiting high yields).

Scenario B: Steam reforming of natural gas to hydrogen – reference case.

2.2. Scope

The geographical scope of the WTW study falls into the European region, with a future outlook of 5–10 years. Therefore, current technologies and practices widely used in Europe are considered.

The reference flow for the comparison of the two hydrogen production scenarios is defined as 100 km distance covered by a H₂-internal combustion engine vehicle on the NEDC (New European Driving Cycle) cycle.

The inventory data collected consists of the lifecycle energy use (both fossil and renewable) and the release of greenhouse gas emissions CO₂, CH₄ and N₂O. Other airborne emissions such as NO_x, SO_x, particulates etc are not considered due to lack of detailed data for the investigated processes/lifecycle steps.

The assessment of each fuel is based on classification of the input/outputs into impact categories and their subsequent evaluation. The following impact categories are considered:

Abiotic depletion (ADP): Important is the use of fossil energy resources. The depletion of other abiotic resources is included in this indicator as well. The reference unit is MJeq.

Cumulative energy demand (CED): The cumulative energy demand of biomass, other renewable, fossil and nuclear energy resources is characterized and summed up with the reference unit MJeq. For agricultural products, the energy content in the harvested biomass is considered for this indicator.

Global warming (GWP): Contribution to the problem of climate change evaluated with the global warming potential. The reference unit is CO₂-eq. CO₂, CH₄ (21 GWP) and N₂O (310 GWP) are considered as greenhouse gases [35].

There is general consensus that *land use change*, the conversion of existing land from its current use to the cultivation of biomass feedstock for biofuel production, may have consequences in the GHG emission balance via the release of carbon stored in the soil as CO₂ emissions when the soil is disturbed. Even though these emissions can dramatically change the results of the LCA assessment, GHG effects of direct and indirect land use change were not taken into account in the present LCA due to the complexity and uncertainty associated with existing models for land use changes. There is still considerable uncertainty and lack of knowledge regarding the loss from or sequestration of carbon in solids due to the change in land use. According to Azapagic and Stichnothe [36], it is also unclear how temperature increase due to climate change might alter farm management practices and what effect that will have on the change of carbon to soils.

2.3. System boundaries and limitations

In order to obtain consistent and comparable results, the steps in the lifecycle of hydrogen that should be taken into account (i.e., system boundaries) need to be carefully defined. For a consistent and robust study, the lifecycle of all investigated hydrogen scenarios is split into two stages:

- **Well-to-Tank (WTT)** stage that includes biomass cultivation and/or resource extraction, transportation to production site, conversion to hydrogen and compression and distribution to fuel station
- **Tank-to-Wheels (TTW)** stage that includes the final combustion of the fuel in the ICE vehicle

Additionally, the WTW considers the lifecycle of all auxiliary processes in each of the above steps, such as electricity generation, natural gas provision, diesel fuel production, fertilizer production etc. The detailed boundaries and processes considered (both main and auxiliary) in the hydrogen scenarios from biomass (Scenarios A1 and A2) and from natural gas (Scenario B) are shown in Fig. 1A & B and Fig. 2, respectively. The main assumptions for each investigated option are reported in detail in the LCI section (Section 3).

The inventory within the WTW study considers all relevant environmental flows according to the attributional modeling principle. Thus the results show the environmental impacts caused by the production processes. The modeling does not consider changes introduced by the extension of the market share of these production processes or increased production of hydrogen. Moreover, the life cycle environmental flows associated with producing capital equipment and facilities are excluded from this study.

3. Lifecycle inventory (LCI)

3.1. Well-to-Tank (WTT)

3.1.1. Scenario A: hydrogen from biomass pyrolysis and bio-oil steam reforming

The lifecycle WTT inventory, in terms of energy requirements (fossil and renewable), total energy efficiency and GHG

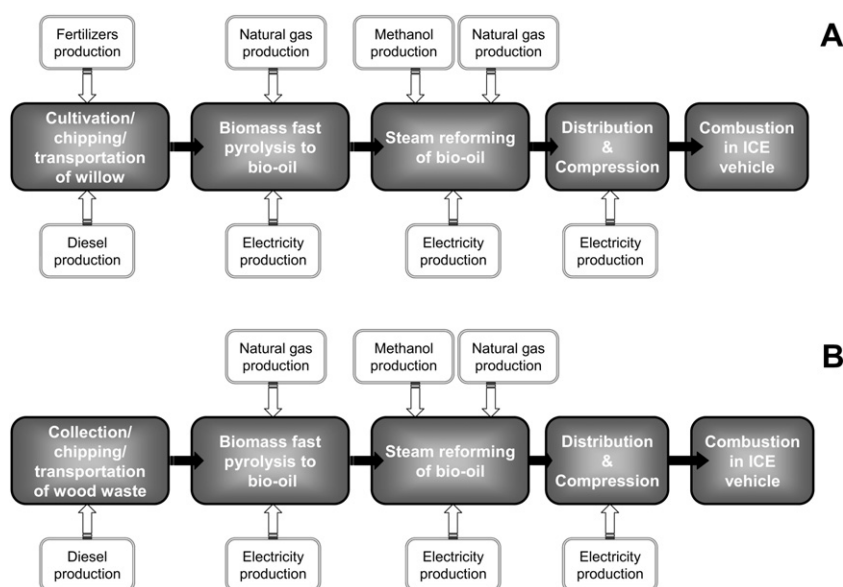


Fig. 1 – System boundaries and processes in the lifecycle of hydrogen from biomass pyrolysis and bio-oil steam reforming for use in H₂-ICE vehicle from (A) willow cultivation; (B) wood waste.

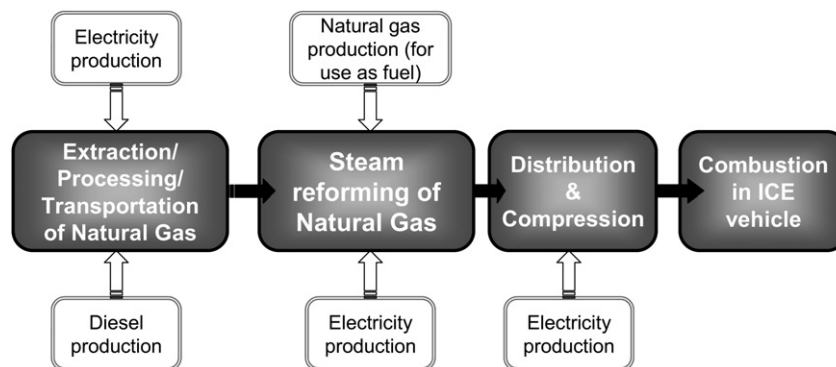


Fig. 2 – System boundaries and processes in the lifecycle of hydrogen produced from natural gas steam reforming for use in H₂-ICE vehicle.

emissions per MJ of produced hydrogen in the hydrogen gas station for each lifecycle step in Scenarios A1 and A2 are tabulated in Table 1A and B, respectively. Detailed analysis of each of these steps is given in the following paragraphs.

3.1.1.1. Provision of biomass feedstock (collection/cultivation, chipping, transport). As already mentioned, two pathways have been considered for the provision of lignocellulosic biomass feedstock for the pyrolysis process: wood waste and dedicated willow plantations. For both feeds, this first lifecycle step includes the collection or cultivation/harvesting of biomass, its chipping and its transportation to the processing plant.

In the case of wood waste, the process chain starts with the collection of the wood residuals. Mass losses of about 0.01 MJ/MJ_{wood} occur during chip making, handling and storage, due to spillage, evaporation of volatiles, respiration and rotting [18]. Energy is expended as diesel fuel for operation of the forestry

machinery. The energy requirement and the associated GHG emissions from the supply of residual woody biomass (including chipping and transport) are derived from the GEMIS database, reported in the GM European WtW study [18]. The average transport distance is assumed to be 50 km. The values include also the energy demand and GHG emissions from the production of diesel used in forestry machinery and transportation trucks.

When willow is used as feedstock, the lifecycle chain additionally includes the cultivation of the biomass. The inputs in the cultivation process comprise sowing, thinning, fertilizer, but mostly harvesting. Farming inputs vary widely, depending on soil quality, yield and the intensiveness of the farming. Data concerning the average EU inputs for wood farming (SRF, willows or poplars) were obtained from the GEMIS database, reported in the GM European WtW study [18]. These include the amount of fertilizers and diesel fuel used for the operation of the farming machinery and their associated

Table 1 – WTW energy use, energy efficiency and GHG emissions of the individual steps and total lifecycle of hydrogen from biomass pyrolysis and bio-oil steam reforming for use in H₂-ICE vehicle from (A) willow cultivation; (B) wood waste.

A		Waste wood collection/ chipping/transportation	Biomass pyrolysis to bio-oil	Steam reforming to H ₂	H ₂ distribution/ compression	Total
Energy use (MJ/MJ H ₂)	Total	0.087	0.837	0.557	0.222	1.704
	Fossil	0.071	0.259	0.467	0.192	0.989
	Renewable	0.016	0.579	0.090	0.030	0.715
Total energy efficiency (energy out/energy in)		92.0%	54.4%	64.2%	81.8%	37.0%
GHG emissions (g/GJ H ₂)	CO ₂	5.280	15.244	19.126	11.700	51.350
	CH ₄	0.000	26.445	24.333	17.999	68.78
	N ₂ O	0.000	0.400	0.569	0.421	1.39
B		Willow cultivation/harvesting/ chipping/transportation	Biomass pyrolysis to bio-oil	Steam reforming to H ₂	H ₂ distribution/ compression	Total
Energy use (MJ/MJ H ₂)	Total	0.124	0.837	0.544	0.222	1.741
	Fossil	0.107	0.259	0.454	0.192	1.025
	Renewable	0.016	0.579	0.090	0.030	0.715
Total energy efficiency (energy out/energy in)		89.0%	54.4%	64.8%	81.8%	36.5%
GHG emissions (g/GJ H ₂)	CO ₂	7.425	15.244	19.125	11.700	53.495
	CH ₄	4.950	26.445	24.333	17.999	73.73
	N ₂ O	36.303	0.400	0.569	0.421	37.69

emissions, as well as the corresponding lifecycle of each of these inputs (e.g., the required energy and emissions for the production of diesel fuel and N-fertilizer). Dry mass losses during chipping and storage, partly from dust and spillage, and partly from respiration, rotting and evaporation of volatiles, were also taken into account (same value as in wood waste). The figures contain also transportation to the processing plant, at an average distance of 50 km.

3.1.1.2. Fast pyrolysis of woody biomass to bio-oil. Bio-oil is produced via the fast pyrolysis of biomass, a high-temperature process in which the feedstock is rapidly heated in the absence of air, vaporises and condenses to a dark brown flowing liquid [4]. The main product, bio-oil, is obtained in yields of up to 80 wt.% on dry feed basis, together with by-products char and gases, which are used within the process to cover the energy and heat load.

Modeling of the pyrolysis process, in order to calculate the energy expenditure and associated emissions for bio-oil production, was based on public process data for the Dynamotive's proprietary BioTherm process [37], which were provided to the University of New Hampshire (Canada) to conduct a study on the technical, environmental and economic feasibility of bio-oil [38]. The data were scaled up for a plant with capacity of 400 tons/day (wet feedstock) [38].

The fast pyrolysis process, shown graphically in Fig. 3, can be subdivided in the following three sections: pre-treatment, reaction and collection of the liquid product in the downstream. In the *pre-treatment* step, the biomass feed is first dried to less than 10% water in order to minimize the water in the product liquid oil. For calculating the heat demand for the drying step, it was assumed that the as-received woody biomass has a moisture content of 45%, which is reduced down to 10% for feeding to the reactor [38]. The required heat for drying is provided by combustion of the char by-product of the pyrolysis reaction. Calculations showed that with a char yield of 21%, all produced char is utilized for covering the heat demands of the drying step [38]. After drying, the biomass has to be

grinded to ~ 2 mm to give small particles and ensure rapid heating and reaction, a factor which is critical for maximizing the liquid yield of the process. Grinding consumes electricity from the grid, derived from the average EU 27-electricity mix. The energy use and GHG emissions associated with the lifecycle of average EU 27-electricity mix production were obtained from the European Life Cycle Database (ELCD II) [39].

In the *reaction* step, a bubbling fluidized bed pyrolysis reactor is used to convert the wood chips to three components: char, condensable and non-condensable gases with an average yield of 21%, 66% and 13% (on dry feed) respectively [38]. The shredded biomass is fed to the reactor, where it is rapidly heated to temperature ~ 450 °C by fluidized sand, in the absence of oxygen. Simultaneously, pre-heated non-oxidizing gas (external inert nitrogen and recycled non-condensable gases) is introduced in the reactor at sufficient linear velocity to act as a fluidization medium for the hot sand bed. The bed is heated indirectly by flue gases in the jacket of the reactor, provided by combustion of part of the non-condensable gaseous products of the reaction and external heat from a natural gas burner. The heat for the pyrolysis is the total heat that must be delivered to the reactor to provide all the sensible, radiation and reaction heat for the process to proceed to completion. The heat of reaction for the fast pyrolysis process is marginally endothermic. According to Dynamotive [37], 2.5 MJ of energy are required in the pyrolysis for the production of 1 kg of bio-oil, 60% of which is provided by the non-condensable gaseous by-product. The net heat required from external sources (natural gas) is 1 MJ/kg bio-oil [37]. Natural gas is assumed to derive from the average EU-27 natural gas mix. The lifecycle of natural gas, including its extraction/processing/transportation to the plant is described in detail in section 3.2.1. The natural gas burner emissions were based on emissions from EPA for natural gas combustion in industrial boilers, obtained from GREET [40].

The products exit the pyrolysis reactor in a gaseous state that includes vapors, aerosols, and a suspension of small solid particles. In the *product recovery* step, the exit stream first passes through a series of cyclones, which separate most of the solid particles from the vapors. The char is removed and collected in a quench system, the gases are then condensed in a quench system, which converts most of the vapor into liquid bio-oil by rapid cooling to below 50 °C using previously made bio-oil as quenching agent [38]. The non-condensable gases (CO_2 , CO , CH_4) enter an electrostatic precipitator, which cleans the gases from residual bio-oil aerosol droplets and particulates, and are then recycled back to the reactor as inert gas for fluidization and to the separate combustion chamber for indirect heating of the sand bed.

Summarizing, the fast pyrolysis process is largely self-sufficient in terms of energy use, due to the integration of the by-products in the actual process. Since it was assumed that both char and non-condensable gases are consumed internally to cover the process energy requirements, the sole product of the process is bio-oil and therefore no allocation was performed and all environmental burdens were attributed to bio-oil. The limited external energy requirements consist of natural gas used as auxiliary fuel for heating the sand bed and electricity used for biomass grinding, electrostatic precipitator, pumps, compressors etc. The inputs for the

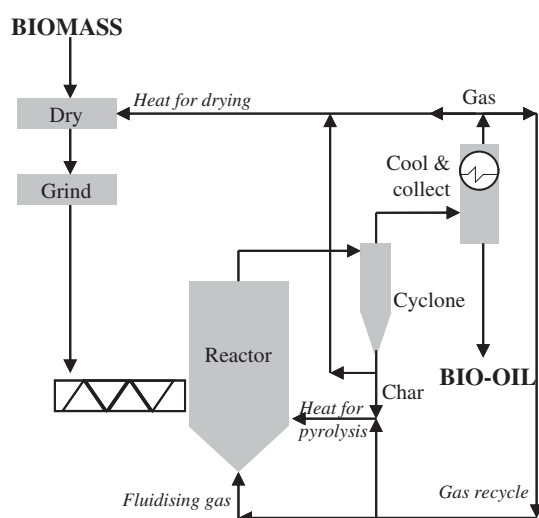


Fig. 3 – Typical flow diagram of a biomass fast pyrolysis process.

sand heating medium and the inert fluidization gas (nitrogen) are minor in terms of primary energy requirements, since they are recycled and therefore their lifecycle was not taken into consideration.

3.1.1.3. Steam reforming of bio-oil to hydrogen. Bio-oil is converted to hydrogen via steam reforming. It is assumed here that the whole fraction of bio-oil is used in a centralized unit, which includes both the pyrolysis and the reforming units. Therefore, no transportation of the bio-oil is assumed. Modeling of the steam reforming process was based on a concept investigated by the National Renewable Energy Laboratory (NREL) [6,9], according to which bio-oil is converted to hydrogen in a fluidized bed reactor in the presence of commercial nickel-based catalysts. A fluidized bed configuration is preferable, as bio-oil steam reforming is limited by extensive coking when fixed-bed units are employed. According to Abedi et al. [9], the use of a fluidized bed reactor greatly increases reforming efficiency and extends the catalyst time-on-stream. Data on the energy use during steam reforming of bio-oil in a fluidized bed reactor, according to the NREL concept, were obtained from Evans et al. [33].

Prior to the reaction, bio-oil is mixed with methanol. The mixing solvent (10 wt.% methanol) reduces viscosity and density and stabilizes the water-insoluble fraction of bio-oil for long-term storage without phase separation [41,42]. It was assumed that conventional methanol derived from natural gas is used. The lifecycle of methanol produced in an EU methanol plant, supplied with natural gas from the EU 27 natural gas mix grid, including resource extraction, conversion and transportation and distribution, was obtained from the GM European WtW study [18].

After mixing, the bio-oil/methanol mixture is introduced to the fluidized bed reformer. A typical flow diagram is shown in Fig. 4. Steam (steam-to-carbon ratio = 2.5) is used as the fluidizing agent and for transferring heat to the reactor. It is assumed that the reaction is run autothermally at 650 °C with an oxygen/carbon ratio of 0.7 and produces CO₂, CO, H₂, H₂O and CH₄ which are cooled and compressed after removing solid particles (i.e. char and ash) from the product gas [6,29,33]. The reaction products exit the reactor and enter a heat exchanger for cooling prior to entering a water gas shift reactor. The water gas shift reactor is necessary in order to enrich the exit stream in hydrogen. The produced synthesis gas is therefore directed to high and low temperature water gas shift reactors in order to convert produced CO to H₂. Hydrogen is separated and purified in a pressure swing adsorption (PSA) unit. A hydrogen yield of 11.9 wt.% from the bio-oil/methanol blend is assumed [33].

The off-gases of the PSA unit, as well as the heat recovered from the reformed gases, are used to produce the necessary steam for the whole process, therefore no external fuel is considered for steam production. The operation of pumps, compressors and air separation units consumes electricity from the grid, derived from the average EU 27-electricity mix. The energy use and GHG emissions associated with the lifecycle of average EU 27-electricity mix production were obtained from the ELCD II database [39].

3.1.1.4. Distribution & compression. Produced hydrogen is assumed to be distributed via pipelines to the filling station at a distance of 50 km. According to the GM European WtW study [18], the pressure of hydrogen in the production plant is adequate for its transportation via pipelines in order to arrive in the filling station with a pressure of 1.5 MPa after the pressure drop and losses in the hydrogen pipeline grid.

For the refueling of the H₂-ICE vehicle, a vehicle tank pressure of 35 MPa is assumed. In this case, the filling station needs to have hydrogen compressed up to about 45 MPa [18]. Compression demands the use of electricity, considered to be equal to 0.018 kWh/MJ H₂ for hydrogen compression from 1.5 MPa to 45 MPa, and leads to hydrogen losses in the order of 2% [18]. Electricity is assumed to be derived from the average EU 27-electricity mix. The energy use and GHG emissions associated with the lifecycle of average EU 27-electricity mix production were obtained from the ELCD II database [39].

3.1.2. Scenario B: hydrogen from natural gas steam reforming—base case

The WTT lifecycle inventory, in terms of energy requirements (fossil and renewable), total energy efficiency and GHG emissions per MJ of produced hydrogen in the hydrogen gas station, for each lifecycle step in Scenario B is tabulated in Table 2. Detailed analysis of each of these steps is given in the following paragraphs.

3.1.2.1. Extraction/processing/transportation of natural gas.

The natural gas production process consists of the natural gas extraction from on-shore and off-shore wells and the natural gas processing stage. The extraction occurs at the well (depth up to 5000 m). From the wells, the natural gas is piped to the natural gas processing plant by gathering pipelines [18]. Processing, carried out at terminals prior to transmission to the pipeline network, is minimal and involves only removal of hydrogen sulfide and drying, resulting thus to SO₂ as the main emission [18]. After processing, natural gas is distributed to the end-customers. The transportation of natural gas from the oil field to the end-customer via pipeline accounts for a large

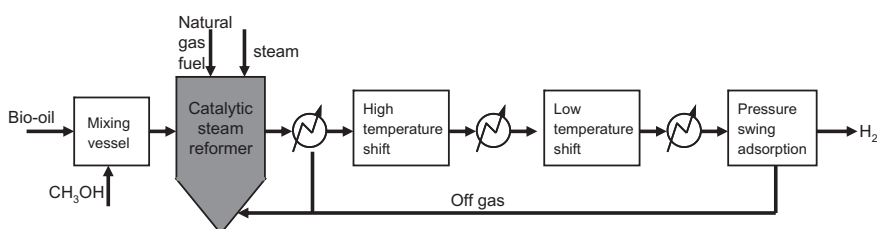


Fig. 4 – Typical flow diagram of a bio-oil steam reforming process.

Table 2 – WTW energy use, energy efficiency and GHG emissions of the individual steps and total lifecycle of hydrogen produced from natural gas steam reforming for use in H₂-ICE vehicle.

		Extraction/processing/ transportation of NG	NG steam reforming to H ₂	H ₂ distribution/ compression	Total
Energy use (MJ/MJ H ₂)	Total	0.147	0.205	0.222	0.575
	Fossil	0.147	0.205	0.192	0.545
	Renewable	0.000	0.000	0.030	0.030
Total energy efficiency (energy out/energy in)		87.2%	83.0%	81.8%	63.5%
GHG emissions (g/GJ H ₂)	CO ₂	6.772	9.764	11.700	28.236
	CH ₄	163.489	20.686	17.999	202.17
	N ₂ O	0.130	0.195	0.421	0.75

part of the energy requirement because of the large distances involved. Pipelines require compression stations at regular intervals, typically powered by a portion of the transported gas. Therefore, the energy requirement depends mainly on the piping distance and the pressure at which the pipeline is operated. Existing pipelines operate at pressures of 6–8 MPa [18]. Combined leakages in the transportation system result in some methane losses (directly emitted to the atmosphere).

As the geographical scope of this WTW study falls into the European region, the origin of the natural gas feedstock for hydrogen production via steam reforming is assumed to be that of the EU-27 average natural gas mix. This mix consists of the domestic EU natural gas production, plus the complimentary imports by countries outside EU. European natural gas production mainly originates from the oil fields in UK, Norway and Netherlands, where natural gas is extracted as a co-product of crude oil, and it is complimented by imports mainly from Russia and Algeria [20]. Data for energy consumption and GHG emissions for the extraction/processing and transportation of the average EU-27 natural gas mix were obtained from the ELCD II database [43]. The data set considers the whole supply chain of natural gas i.e. exploration, production, processing (e.g. desulphurization) and in case of LNG import, liquification/regasification of LNG, the long distance transport and the regional distribution to the final consumer. Losses occurring during transportation via pipeline or vessel are included.

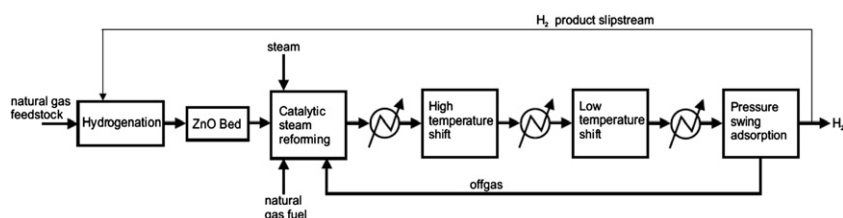
3.1.2.2. Steam reforming of natural gas to hydrogen. Hydrogen production via catalytic steam reforming of natural gas is a mature technology and is the route by which most hydrogen is made today. Modeling of the natural gas steam reforming process in the present WTW study was based on data from Spath et al. [17], which were based on previous SRI data for a hydrogen plant modified to include a low temperature water gas shift reactor. The modeled plant has a hydrogen

production capacity of 1.5 million Nm³/day (typical size of centralized steam reformers) [17].

A typical flow diagram of a steam reforming process is shown in Fig. 5. Natural gas is first pretreated in a hydrogenation vessel in order to convert any sulfur compounds to H₂S, which is further removed over a ZnO bed. This step requires a small amount of hydrogen, which is recycled from the product stream. After pre-treatment, natural gas is mixed with steam and is fed to the reforming reactor. The produced synthesis gas is then directed to high and low temperature water gas shift reactors in order to convert produced CO to H₂. Hydrogen is separated and purified in a pressure swing adsorption (PSA) unit. The PSA unit produces an off-gas, consisting of CO₂, H₂, CH₄, CO and N₂, which is used to fuel the reformer. According to Spath et al. [17], an additional small amount of natural gas (4.4 wt.% of the total reformer fuel requirement) is needed to supply the balance of the reformer duty. The natural gas burner emissions were based on emissions from EPA for natural gas combustion in industrial boilers, obtained from GREET [40]. The steam reforming process is a net steam producer, which is exported for use by some other process or facility. The operation of pumps and compressors consumes electricity from the grid, derived from the average EU 27-electricity mix. The energy use and GHG emissions associated with the lifecycle of average EU 27-electricity mix production were obtained from the ELCD II database [39].

To summarize, the energy consumption of the process consists basically of the natural gas used as fuel for providing heat to the reformer (plus the energy for its extraction and transportation), electricity for operation of pumps (plus the energy for its extraction and transportation) and the energy efficiency losses.

3.1.2.3. Distribution & compression. The assumptions made for the distribution of hydrogen from natural gas to the filling

**Fig. 5 – Typical flow diagram of a natural gas steam reforming process [17].**

station and its compression for the refueling of the H₂-ICE vehicle are identical to those of hydrogen from biomass and are not repeated for brevity. Details can be found in Paragraph 3.1.1.

3.2. Tank-to-Wheel (TTW)

The TTW stage of all investigated hydrogen options is common and involves the combustion of hydrogen in a conventional internal combustion engine vehicle for covering a distance of 100 km in the New European Driving Cycle (NEDC). Data for the energy use in a H₂-ICE vehicle were obtained from the CONCAWE-JRC-Eucar WTW study [20], where fuel consumption and GHG emissions were simulated according to the standard European driving cycle with an open source vehicle simulation tool. The simulation was based on a common, “virtual” vehicle, representing a typical European compact size 5-seater sedan, comparable to e.g. a VW Golf or others in that class [20]. According to the above assumptions, the energy consumption for covering a distance of 100 km was calculated equal to 167.5 MJ H₂/100 km. This value is used in the present study to calculate the WTW energy use and emission of the different hydrogen production pathways. The combustion of hydrogen in the vehicle is “clean” and releases no greenhouse gas emissions.

4. Lifecycle impact assessment (LCIA) & results interpretation

4.1. Abiotic depletion

As described earlier, the abiotic depletion impact category includes, in the case of hydrogen, the fossil energy demand during the whole WTW lifecycle of hydrogen produced via the different processes. Fig. 6 illustrates the impact of these hydrogen WTW scenarios to the depletion of fossil energy resources, with the relative contributions from each different phase in hydrogen’s lifecycle.

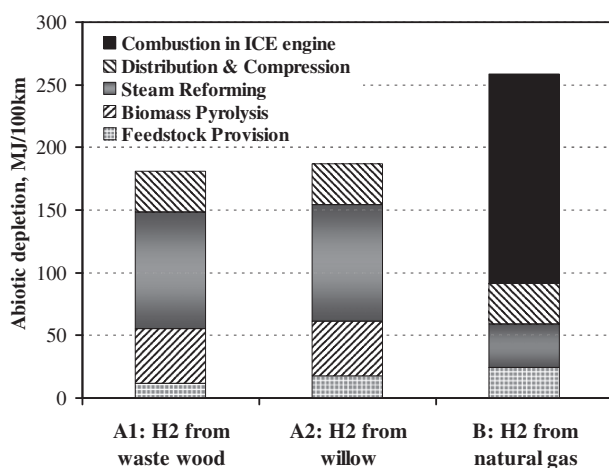


Fig. 6 – Impact of the different hydrogen scenarios’ to abiotic depletion impact category.

It can be clearly deduced from this figure that the biomass-derived hydrogen, either from wood waste or dedicated willow cultivation, offers considerable reductions in WTW fossil energy use compared to conventional hydrogen from steam reforming. This is mainly due to the renewable nature of the biomass feedstock, and as a consequence of the fuel itself, which results in no fossil energy consumption during the combustion step in the H₂-ICE vehicle. Therefore, for the biomass-derived hydrogen the WTT results basically coincide with the WTW analysis for the fossil resources depletion category, since the TTW contribution is nil.

It should however be noted that when comparing the consumption of fossil energy in the processes prior vehicle use (WTT consumption), the production of biomass-derived hydrogen with the current technology, irrespective of the biomass feedstock, is more energy demanding than conventional hydrogen production from natural gas. Although the feedstock provision step is more energy-consuming for methane extraction compared to willow cultivation or wood waste collection, the lifecycle of hydrogen from biomass inherently includes an extra process, biomass pyrolysis to bio-oil, which aggravates the fossil energy balance. Still, the steam reforming process which is common to all hydrogen production pathways demands higher consumption of fossil energy when biomass is the feed compared to natural gas.

In order to further investigate the origin of the higher energy demand in biomass steam reforming as opposed to natural gas reforming, the source of energy consumption in the two processes is shown in Fig. 7. It becomes evident that the origin of this significant difference is the use of methanol in bio-oil steam reforming, which is added to stabilize the bio-oil and reduce extensive coking thus extending the catalyst lifetime, and the use of electricity, which is much higher in bio-oil reforming. If biomass-derived methanol would be used, the energy consumption for its production, as well as its renewable nature, would minimize the contribution of methanol in the energy balance. In this study, it was assumed that conventional methanol from natural gas is used. The fossil energy consumption in the lifecycle of EU fossil methanol was obtained from the GM European WtW study [18], where it was calculated to be equal to 0.62 MJ/MJ methanol. The lifecycle of methanol produced from farmed lignocellulosic was calculated in the CONCAWE-JRC-Eucar WTW

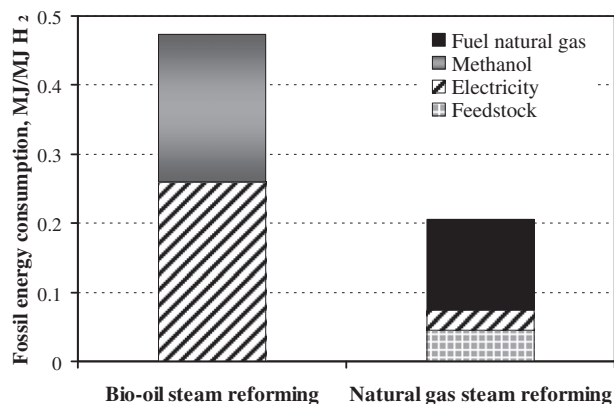


Fig. 7 – Analysis of fossil energy consumption in bio-oil and natural gas steam reforming processes.

study [20] as 0.06 MJ/MJ methanol, an order of magnitude lower than fossil methanol. Thus, it is clear that “renewable” methanol would significantly improve the energy balance of hydrogen production from bio-oil. Still electricity in bio-oil steam reforming surpasses the energy use in natural gas steam reforming. This occurs as bio-oil reforming is assumed to run autothermally, i.e. with the presence of oxygen, which is obtained from Air Separation Units that operate and thus consume a lot of electricity. Further improvement of the fossil energy balance for bio-oil reforming can occur if electricity from renewable energy sources is employed. However, this scenario is not realistic for the time frame of this WTW study, which has a future outlook of 5–10 years.

As means of sensitivity analysis, the possibility of the natural gas reforming plant to include carbon capture and storage was also investigated. Such an addition would aggravate the fossil energy use in the plant with about 16% [20]. The results of the sensitivity analysis showed that even with addition of carbon capture and storage, the steam reforming of natural gas would still be less energy-consuming than bio-oil reforming.

Comparing the two lignocellulosic biomass provision pathways, it can be deduced that the cultivation of willow aggravates the use of fossil resources by ~5%, as opposed to the use of forestry and agricultural waste. However, given the overall differences in energy use between conventional and bio-hydrogen, this increase is not significant enough to alter the generally favorable trend for biomass-derived hydrogen.

4.2. Cumulative energy demand

The cumulative energy demand includes the total energy demand (both fossil and renewable) during the whole lifecycle of hydrogen produced via the different processes. The total energy consumption of the three investigated scenarios, with the relative contributions from each different phase in hydrogen's life, is presented in Fig. 8.

When renewable energy is also considered, the energy balance becomes now unfavorable for biomass-derived hydrogen, compared to fossil energy demand. Whereas the fossil hydrogen's balance remains the same as no renewable

energy is consumed in its lifecycle, the bio-derived hydrogen's energy consumption is significantly aggravated due to the inclusion of the renewable energy consumed in the form of the bio-oil feedstock. This is an indication of the lower efficiency of the conversion of bio-oil to hydrogen compared to the conversion of natural gas and it basically reflects the radically different nature of the two feedstocks. It should be born in mind that bio-oil has a H/C atomic ratio of about 1.3 [33], while natural gas has a ratio of 4. The difference is even bigger if the initial H/C ratio of biomass is considered. It is thus inevitable that more biomass than natural gas has to be consumed for the production of the same amount of hydrogen, given also the two-step conversion process.

Concerning the use of willow cultivation versus wood waste, as discussed earlier, no significant differences can be observed.

4.3. Global warming

The contribution of hydrogen's lifecycle to global warming and climate change due to emissions of CO₂, CH₄ and N₂O is presented in Fig. 9, with the relative contributions from each different phase in the investigated lifecycles. It should be noted that only fossil CO₂ was taken into account.

The biomass-derived hydrogen scenarios exhibit major reduction in greenhouse gas emissions. More specifically, when hydrogen is produced from willow cultivation the reduction is in the order of 37%, whereas when the use of wood waste is considered as feedstock, this reduction increases to 50%. The analysis of the relative contribution of each lifecycle step in the GHG emissions balance points out that the origin of the increased emissions for conventional hydrogen is the steam reforming plant. The production of hydrogen from natural gas involves the removal of the carbon from methane, which is emitted in the off-gases as CO₂. The same of course applies when bio-oil is used as feedstock. However, due to the renewable nature of biomass, and as a consequence of bio-oil, the amount of CO₂ released from the reforming plant is considered to be adsorbed by biomass during its cultivation and therefore is not accounted for in the GHG emissions balance. Capture and sequestration of CO₂

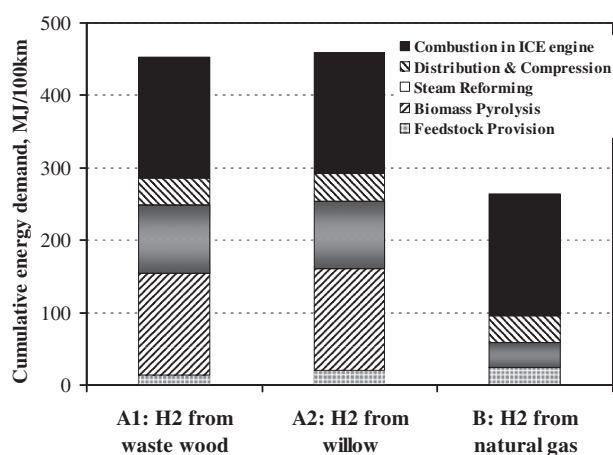


Fig. 8 – Impact of the different hydrogen scenarios' to cumulative energy demand impact category.

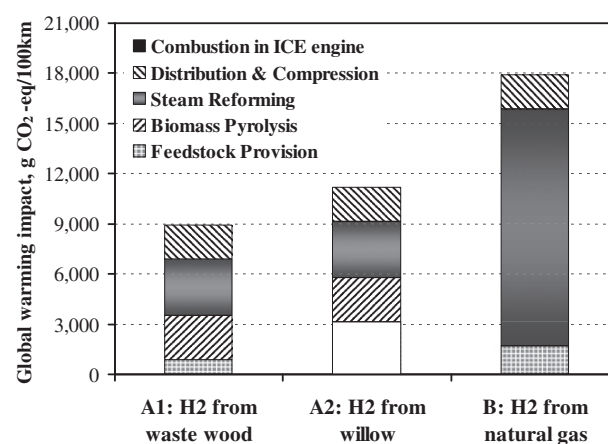


Fig. 9 – Impact of the different hydrogen scenarios' to global warming impact category.

emitted during the steam reforming of natural gas would significantly improve the GHG balance for fossil hydrogen. Data show that GHG emissions in the steam reforming plant would be reduced by ~84% [20]. A rough calculation with this number showed that in the case of carbon capture and sequestration in the steam reforming plant, the GHG emissions for fossil hydrogen would be ~40% less than the bio-hydrogen equivalent.

Concerning the two lignocellulosic biomass provision pathways, global warming is the only environmental impact category where significant differences are evident. The use of cultivated biomass feedstock increases GHG emissions by ~20%. These high emissions are related to the release of N₂O, a gas with extremely high global warming potential, emitted both from the soil during the crops growth step and from the production of fertilizers. Reduction in the amount of N-fertilizers employed in the field, as well as modern agriculture methods, would in the long-term decrease the GHG balance for cultivated biomass.

5. Conclusions

The WTW analysis of hydrogen production from biomass via pyrolysis and subsequent steam reforming of bio-oil, and hydrogen production from natural gas, for use in internal combustion engine vehicle, yielded interesting results. Comparison in terms of fossil energy consumption in the lifecycles of biomass-derived and conventional hydrogen indicates that bio-hydrogen, either from wood waste or dedicated willow cultivation, offers considerable reductions compared to conventional hydrogen from natural gas. This is mainly due to the renewable nature of the biomass feedstock, and as a consequence of the fuel itself, which results in no fossil energy consumption during the combustion step in the H₂-ICE vehicle. Still, the processes prior to combustion are more energy demanding in the case of hydrogen from biomass. The origin of the higher energy consumption is in the bio-oil reforming step, due to the use of methanol, which is added to stabilize the bio-oil and reduce extensive coking, and the increased electricity demand. Further improvement of the fossil energy balance for bio-oil reforming can occur if methanol and electricity produced from renewable energy sources are employed.

Even though bio-hydrogen production is in total more energy intensive than natural gas reforming, the overall benefit of using a CO₂ neutral renewable feedstock for the production of hydrogen as a transportation fuel is unquestionable. The biomass-derived hydrogen scenarios exhibit major reduction in greenhouse gas emissions. More specifically, when hydrogen is produced from willow cultivation the reduction is in the order of 37%, whereas when the use of wood waste is considered as feedstock, this reduction increases to 50%. Moreover, a significant difference in GHG emissions was found between the two different biomass sources. The use of cultivated biomass feedstock increases GHG emissions by ~20%, due to N₂O emissions, emitted both from the soil during the crops growth step and the production of fertilizers.

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